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Spatial Differentiation in the Characterisation of Photochemical Ozone Formation The EDIP2003 Methodology

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Abstract

Background, Aims and Scope. In the life cycle of a product, emissions take place at many different locations. The location of the source and its surrounding conditions influence the fate of the emitted pollutant and the subsequent exposure it causes. This source of variation is normally neglected in Life Cycle Impact Assessment (LCIA), although it is well known that the impacts predicted by site-generic LCIA in some cases differ significantly from the actual impacts. Environmental impacts of photochemical ozone (ground-level ozone) depend on parameters with a considerable geographical variability (like emission patterns and population densities). A spatially differentiated characterisation model thus seems relevant.

Methods and Results. The European RAINS model is applied for calculation of site-dependent characterisation factors for Non-Methane Volatile Organic Compounds (NMVOCs) and nitrogen oxides (NO_x) for 41 countries or regions within Europe, and compatible characterisation factors for carbon monoxide (CO) are developed based on expert judgement. These factors are presented for three emission years (1990, 1995 and 2010), and they address human health impacts and vegetation impacts in two separate impacts categories, derived from AOT40 and AOT60 values respectively. Compatible site-generic characterisation factors for NMVOC, NO_x , CO and methane (CH_4) are calculated as emission-weighted European averages to be applied on emissions for which the location is unknown. The site-generic and site-dependent characterisation factors are part of the EDIP2003 LCIA methodology. The factors are applied in a specific case study, and it is demonstrated how the inclusion of spatial differentiation may alter the results of the photochemical ozone characterisation of life cycle impact assessment.

Discussion and Conclusions. Compared to traditional midpoint characterisation modelling, this novel approach is spatially resolved and comprises a larger part of the cause-effect chain including exposure assessment and exceeding of threshold values. This positions it closer to endpoint modelling and makes the results easier to interpret. In addition, the developed model allows inclusion of the contributions from NO_x , which are ne-

glected when applying the traditional approaches based on Photochemical Ozone Creation Potentials (POCPs). The variation in site-dependent characterisation factors is far larger than the variation in POCP factors. It thus seems more important to represent the spatially determined variation in exposure than the difference in POCP among the substances.

Keywords: Life cycle impact assessment, photochemical ozone formation, vegetation impact, human health impact, spatial differentiation, site-dependent, site-generic, characterisation

1 Background, Aims and Scope

Ozone formation takes place when solvents and other volatile organic compounds (VOCs) and carbon monoxide (CO), released into the atmosphere, are degraded, and in reactions with nitrogen oxides (NO_x), initiated by sunlight, form ozone. The nitrogen oxides are not consumed during the ozone formation, but have a catalyst-like function. The reactions take place in the troposphere, the lower 8–12 km of the atmosphere, where they are the primary source of ozone. A distinction is made between the ground-level ozone formation, which is mainly driven by NO_x and NMVOCs, and the ozone formation in the free troposphere which is governed by CO and methane (CH_4). The ground level ozone has a residence time of a few hours but it is being replenished by the ozone in the free troposphere, where the residence time of ozone is several weeks. In the free troposphere, ozone is thus widely dispersed, and ozone measured at a particular location may have arisen from NMVOC and NO_x emissions many hundreds of kilometres away (Finlayson-Pitts and Pitts 1986, Seinfeld and Pandis 1997). Due to its high reactivity, ozone attacks organic substances present in plants and animals or materials exposed to air. This leads to an increased frequency of humans with problems in the respiratory tract during periods of photochemical smog in cities, and where the tropospheric concentration of ozone is increasing, it may cause stress to vegetation leading to substantial losses in agricultural yields (Fowler et al. 1997).

Existing characterisation factors for photochemical ozone formation in European methods are based on the Photochemical Ozone Creation Potential (POCP) of substances (Derwent and Jenkin 1990, Derwent et al. 1996, Derwent et al. 1998), or possibly their Maximum Incremental Reactivity (MIR) (Carter et al. 1997). Both express the short-term contribution to ozone formation. Ozone formation is strongly dependent on local conditions like the simultaneous presence of other NMVOCs and NO_x , and the solar radiation intensity. All these may vary strongly from location to location. This is why a preliminary spatial differentiation was introduced in the EDIP97 life cycle impact assessment (LCIA) methodology through the distinction between emissions occurring in regions with low and high background levels of NO_x (Hauschild and Wenzel 1998b). This preliminary approach is in the EDIP2003 methodology replaced by an approach with site-dependent characterisation factors for individual European countries based on sophisticated modelling of photochemical ozone formation.

Both POCP and MIR have been developed for ranking of NMVOCs according to their ozone formation potential in the presence of NO_x which is considered a prerequisite. Due to the lack of a POCP factor for NO_x , its contribution to ozone formation is neglected when traditional characterisation factors are applied, and this is a serious flaw. The sophisticated model used to calculate the EDIP2003 factors allowed us to derive site-dependent factors for NO_x as well.

This paper reports on the development of the site-dependent EDIP2003 factors for ozone formation, presents the procedure for how to apply them in LCIA and illustrates their use with an example.

The use of site-dependent characterisation has been topical right from the start, but its acceptance in LCIA is growing. This process is also visible in the development in the position of Helias Udo de Haes over the years from a sceptical opponent to a cautious advocate of site-dependency and, even broader, situation-dependency in LCIA. Helias Udo de Haes at the same time played an important role in bringing the discussion forward on this topic. In a column elsewhere in this issue, the authors give their recollection of the development (Potting and Hauschild 2006).

2 Methods and Results

The EDIP2003 characterisation factors for photochemical ozone formation were developed using the RAINS model (Regional Air Pollution Information and Simulation). The RAINS model is an integrated assessment model that combines information on regional emission levels with information on long-range atmospheric transport in order to estimate patterns of deposition and concentration for comparison with critical loads and thresholds for acidification, eutrophication and tropospheric ozone formation (Amann et al. 1999, Amann and Lutz, 2000, Schöpp et al. 1999). One of the main objectives with the RAINS model is to establish the relationship between the country of an emission and the impact on its deposition areas, and this model was therefore seen as an obvious choice for establishing spatially differentiated characterisation factors for photochemical ozone formation in Europe.

The atmospheric chemistry behind the tropospheric ozone formation is quite complex involving a high number of interacting reactions between many different compounds. The calculation of POCPs thus involved highly complex, mechanistic models, but it will not be feasible to integrate them with an assessment model where source-receptor relationships must also be modelled well. For the modelling of ozone formation within the parcels of air moving from any European grid cell to any other European grid cell, RAINS therefore applies a computationally efficient meta-model which has been statistically derived from a mechanistic model of the highly complex reaction schemes behind the formation of ozone and other photo-oxidants.

The meta model expresses the long-term mean ozone concentration in each grid cell j as a function of the annual NMVOC emission v_i and the annual NO_x emission n_i from emitter country i and the effective NO_x emission en_j experienced in grid cell j over the period considered (en_j is corrected for exchange with the free troposphere) (Eq. 1):

$$\overline{[\text{O}_3]} = k_j + \sum_{i=1}^M (a_{ij}v_i + b_{ij}n_i + c_{ij}n_i^2) + \alpha_j \overline{en_j}^2 + \overline{en_j} \sum_{i=1}^M d_{ij}v_i \quad (1)$$

Where:

- M is the number of emitter countries considered
- k_j includes the effects of background concentrations of O_3 and its precursors and natural NMVOC emissions
- $a_{ij}v_i$ provides the linear country-to-grid contribution from NMVOC emissions in country i (allowing for meteorological effects)
- $b_{ij}n_i$ provides the linear country-to-grid contribution from NO_x emissions in country i (allowing for meteorological effects)
- $c_{ij}n_i^2$ serves as a correction term to allow for non-linearities occurring close to countries with very high NO_x emissions
- $\alpha_j en_j^2$ represents the average non-linearity in the O_3/NO_x relationship experienced over the period considered along the trajectories arriving at grid cell j and any non-linear effects local to grid cell j
- $d_{ij}en_jv_i$ allows for interactions between NO_x and NMVOCs along the trajectories

The coefficients a_{ij} , b_{ij} , c_{ij} , d_{ij} and α_j are estimated by linear regression using n_i , v_i and en_j as variables in the more complex mechanistic model (Heyes et al. 1997a).

The performance of the meta model for ozone formation has been validated by comparing the calculated exposure of vegetation in terms of AOT40 (see definition below) with the results of the complex mechanistic model from which it is derived, in Heyes et al. (1996). Over a realistic range of emissions, it was found to give a very good performance with results on mean ozone concentrations typically deviating less than 1.3% from the full model.

The full model takes into account differences in the composition of the NMVOC emissions for different sectors. Such differences are not included in the meta model and this is a

source of deviation from the results of the full model. To counter this deviation, substance-specific efficiency factors may be applied when the emission of individual NMVOCs is known. The efficiency factors are listed in Annex 1 of the supporting material.

2.1 Calculation of characterisation factors

RAINS can be used to model impacts on human health and ecosystem health (vegetation) through increased exposure to ozone as a result of emissions of NMVOCs or NO_x. Human beings and vegetation have different sensitivity and thresholds to ozone exposure, and their impacts are therefore modelled separately. The impact on man-made materials from ozone is not modelled explicitly by RAINS, but may be considered covered implicitly by the exposure of humans. After all, the geographical distribution of man-made materials to a large extent follows the distribution of humans.

Two (sub) categories of photochemical ozone formation are thus defined, one representing the exposure of human beings and materials, the other representing the exposure of vegetation above their respective thresholds.

The characterisation factor for vegetation exposure to photochemical ozone is calculated as the Accumulated exposure to Ozone above Threshold (AOT40) multiplied by the area of the vegetation (semi natural vegetation and crops) which is exposed. AOT40 is defined as the accumulated dose over a threshold of 40 ppb (ppb=parts per billion: 1 ppb=2 µg/m³), i.e. the sum of the differences between the hourly mean ozone concentration (in ppb) and 40 ppb for each hour when the concentration exceeds 40 ppb, accumulated during daylight hours. The threshold of 40 ppb is chosen as an exposure level below which no or only small effects occur (Kärenlampi and Skarby 1996). The long-term critical level of ozone for crops is the concentration of pollutants in the atmosphere above which adverse effects occur on sensitive crop plants according to present knowledge. This critical level was derived experimentally using open-top chambers to expose field-grown wheat to ozone. The data from spring wheat experiments in 6 countries, 10 seasons and with 10 cultivars were used to derive an exposure response relationship with an r² of 0.88. The AOT40 associated with a 5% reduction in yield, 3 ppm·hours, was accepted as the critical level of ozone for yield reduction in crops (Fuhrer et al. 1997). The critical level of ozone for natural vegetation has been set to the same value. The characterisation factor is calculated as the product of the area of vegetation exposed above the threshold of 40 ppb (m²), the annual duration of the exposure above the threshold (hours), and the extent to which the threshold concentration is exceeded (ppm). The unit of the characterisation factor for vegetation is thus m²·ppm·hours/g substance emitted.

The characterisation factor for human exposure to photochemical ozone is calculated based on the AOT60 multiplied by the number of persons exposed, i.e. the product of the number of persons exposed above the threshold of 60 ppb for chronic effects (pers), the annual duration of the exposure above the threshold (hours), and the extent to which

the threshold concentration is exceeded (ppm). No official threshold for chronic exposure of humans to ozone has been established. Instead, the threshold of 60 ppb is chosen as the long-term environmental objective for a maximum eight-hour average concentration under the EU ozone strategy proposed by the World Health Organisation, WHO (WHO 1987). The unit of the characterisation factor for human exposure is thus pers·ppm-hours/g substance emitted.

Considering that the units for both (sub) categories have an immediate meaning to the person interpreting the results, it is chosen to maintain them rather than express the impacts in terms of a reference substance as is done with the POCPs (C₂H₄ equivalents).

The exposure indices are determined analytically by differentiation of Eq. 1, and for the vegetation in grid cell j, the exposure becomes (Eq. 2):

$$AOT40_j = k_j + \sum_{i=1}^M (a_{ij}v_i + b_{ij}n_i + c_{ij}n_i^2) + \alpha_j \bar{en}_j^2 + \bar{en}_j \sum_{i=1}^M d_{ij}v_i \quad (2)$$

Even though the long-term mean ozone concentration |O₃| expressed by equation (1), and the exposure index AOT40 expressed by Eq. 2, are quite different entities, a very similar form of expression gave the best statistical fit, and the information contained in the parameters of the two equations is analogous, except the variable k_j, which in Eq. 2 includes the effects of background concentration of O₃, its precursors and natural emissions as well as the effect of the threshold value (40 ppb) on the accumulated exposure. The other variables and parameters of Eq. 2 are explained under Eq. 1.

The AOT60 indicator can be calculated analogously.

2.2 Site-dependent characterisation factors

The site-dependent characterisation factors are calculated by multiplying the modelled exposure indices and the mapped areas of vegetation, respectively the number of people exposed, within grid cells and sum the results for 41 countries or regions within Europe. For each country, the site-dependent characterisation factors relate an emission to the ozone exposure and impact on vegetation or human beings within its deposition areas. The site-dependent characterisation factors are shown in Table 1.

The site-dependent characterisation factors in Table 1 show a variation between countries of up to three orders of magnitude for human exposure, with the highest factors for the most densely populated regions. In comparison, the variation is smaller among the factors for vegetation exposure because vegetation is more evenly distributed over Europe than population. The factors depend on the emission patterns of the countries and therefore show some variation in time, as illustrated by Table 1. The underlying per country emissions of non methane VOCs and NO_x are based on the inventories of the UN ECE for 1990 and 1995 while the 2010 emission scenario is estimated based on the current

Table 1: Site-dependent and site-generic characterisation factors for exposure of vegetation (crops and seminatural) or human beings (and man-made materials) to photochemical ozone expressed as cumulative exposure of vegetation above 40 ppb ($m^2\text{-ppm-hours/g}$) and exposure of humans above 60 ppb (pers·ppm-hours/g) respectively. Factors calculated for the emission patterns of 1990 and 1995 and the expected emission pattern of 2010

	Site-dependent characterisation factors											
	Vegetation						Humans					
	NO _x			NMVOC (also proxy for CO)			NO _x			NMVOC (also proxy for CO)		
	1990	1995	2010	1990	1995	2010	1990	1995	2010	1990	1995	2010
Albania	1.1	1.1	1.1	0.19	0.19	0.19	9.0E-06	9.4E-06	2.0E-06	1.7E-06	4.0E-06	9.9E-07
Austria	3.0	3.0	3.0	0.49	0.49	0.45	8.0E-05	7.0E-05	3.7E-05	8.6E-05	4.7E-05	5.0E-05
Belarus	1.6	1.6	1.6	0.40	0.40	0.35	4.4E-06	2.5E-06	8.5E-07	7.4E-06	7.2E-09	3.0E-06
Belgium	1.6	1.6	1.5	1.1	1.1	1.1	4.2E-04	3.8E-04	3.2E-04	2.9E-04	2.2E-04	2.0E-04
Bosnia/Herzegovina	1.6	1.6	1.5	0.22	0.22	0.20	1.3E-05	1.3E-05	3.2E-06	3.8E-06	3.5E-05	2.6E-06
Bulgaria	1.4	1.4	1.4	0.29	0.29	0.28	2.4E-06	2.2E-06	-2.4E-07	6.4E-06	2.2E-06	2.8E-06
Croatia	2.4	2.4	2.3	0.31	0.31	0.29	3.8E-05	3.8E-05	1.6E-05	2.8E-05	1.2E-04	2.1E-05
Czech Republic	2.4	2.4	2.3	0.91	0.91	0.86	2.3E-04	2.1E-04	1.7E-04	1.5E-04	6.2E-07	9.0E-05
Denmark	1.5	1.5	1.4	0.76	0.76	0.66	4.0E-05	3.4E-05	2.2E-05	6.2E-05	2.7E-05	3.8E-05
Estonia	0.17	0.16	0.13	0.62	0.62	0.50	1.4E-06	1.2E-06	6.4E-08	3.7E-07	9.4E-06	3.7E-07
Finland	0.41	0.38	0.33	0.29	0.29	0.25	1.6E-06	8.5E-07	1.7E-07	1.8E-07	5.2E-07	1.0E-07
France	3.4	3.4	3.3	0.88	0.87	0.75	2.4E-04	2.2E-04	1.6E-04	1.5E-04	1.2E-04	7.6E-05
Germany	2.0	2.0	1.9	1.4	1.3	1.2	3.6E-04	3.3E-04	2.7E-04	2.4E-04	1.9E-04	1.4E-04
Greece	0.54	0.55	0.53	0.14	0.14	0.13	1.9E-05	1.9E-05	1.4E-05	1.6E-05	1.1E-05	1.3E-05
Hungary	4.4	4.3	4.3	0.76	0.76	0.73	3.6E-05	3.0E-05	1.2E-05	5.2E-05	2.4E-05	3.1E-05
Ireland	0.21	0.21	0.20	0.068	0.065	0.051	1.5E-05	1.2E-05	8.2E-06	1.5E-05	8.5E-06	9.7E-06
Italy	1.5	1.5	1.5	0.71	0.71	0.63	2.0E-04	2.0E-04	1.4E-04	1.2E-04	1.0E-04	6.6E-05
Latvia	0.41	0.40	0.37	0.26	0.26	0.23	3.8E-06	2.5E-06	1.7E-06	1.0E-06	1.0E-06	1.0E-06
Lithuania	1.1	1.1	1.1	0.57	0.57	0.52	5.7E-06	4.2E-06	1.2E-06	3.4E-06	1.3E-06	1.4E-06
Luxemburg	0.81	0.81	0.79	0.10	0.10	0.10	1.2E-04	1.1E-04	8.6E-05	6.6E-05	5.8E-05	4.9E-05
Macedonia	0.51	0.51	0.50	0.19	0.19	0.19	4.1E-06	4.3E-06	-	-	3.8E-05	-
Moldova	0.69	0.69	0.68	0.54	0.54	0.52	2.5E-06	1.5E-06	-4.9E-07	1.5E-06	1.8E-06	-8.0E-08
Netherlands	0.85	0.83	0.79	0.94	0.94	0.92	2.5E-04	2.3E-04	1.8E-04	1.9E-04	1.3E-04	1.4E-04
Norway	0.26	0.25	0.21	0.10	0.10	0.078	3.6E-06	2.1E-06	6.5E-07	1.9E-06	1.5E-06	9.5E-07
Poland	2.6	2.5	2.5	1.2	1.2	1.0	1.2E-04	1.1E-04	8.6E-05	1.1E-04	6.9E-05	6.1E-05
Portugal	3.5	3.5	3.4	1.0	1.1	1.0	1.3E-04	1.3E-04	9.7E-05	6.7E-05	6.7E-05	4.4E-05
Romania	2.1	2.1	2.1	0.29	0.29	0.28	7.0E-06	5.0E-06	2.2E-08	1.3E-05	5.0E-06	5.7E-06
Russia-Kaliningrad	0.21	0.21	0.21	0	0	0	4.4E-06	3.6E-06	2.4E-06	8.3E-07	4.7E-06	8.4E-07
Remaining Russia	0.91	0.90	0.88	0.17	0.17	0.14	2.4E-06	2.1E-06	1.0E-06	1.8E-06	2.9E-06	5.3E-07
Russia-St.Petersburg	0.035	0.023	0.007	0	0	0	5.9E-07	5.0E-07	2.0E-07	-1.1E-07	2.9E-05	-
Slovakia	3.4	3.4	3.4	0.72	0.72	0.71	6.3E-05	5.6E-05	4.0E-05	5.5E-05	1.5E-06	3.2E-05
Slovenia	1.2	1.2	1.2	0.25	0.25	0.22	2.6E-05	2.6E-05	1.4E-05	1.6E-05	2.7E-06	1.3E-05
Spain	2.3	2.3	2.2	0.63	0.64	0.56	4.8E-05	4.6E-05	2.4E-05	2.7E-05	2.4E-05	1.4E-05
Sweden	1.0	1.0	0.89	0.36	0.37	0.31	1.5E-05	1.2E-05	6.0E-06	1.4E-05	8.3E-06	9.2E-06
Switzerland	2.3	2.2	2.2	0.45	0.45	0.40	1.1E-04	9.8E-05	4.8E-05	1.0E-04	6.1E-05	5.2E-05
Ukraine	2.0	2.0	2.0	0.68	0.67	0.59	5.6E-06	3.7E-06	5.0E-07	2.3E-05	2.1E-07	8.9E-06
United Kingdom	1.0	1.0	1.0	1.3	1.3	1.2	1.1E-04	9.9E-05	8.5E-05	9.5E-05	6.0E-05	9.4E-05
Yugoslavia	1.6	1.6	1.6	0.21	0.21	0.21	3.4E-06	2.2E-06	-1.5E-06	2.9E-06	1.4E-05	1.7E-06
Atlantic Ocean	0.52	0.52	0.51	0.036	0.036	0.036	1.4E-05	1.4E-05	8.6E-06	-	-	-
Baltic Sea	0.54	0.53	0.52	0.038	0.038	0.022	1.9E-06	1.5E-06	7.4E-07	-	-	-
North Sea	1.2	1.1	1.1	0.20	0.21	0.17	8.7E-05	7.8E-05	5.8E-05	-	-	-
	Site-generic characterisation factors											
	Vegetation						Humans					
	NO _x			VOC (also proxy for CO)			NO _x			VOC (also proxy for CO)		
	1990	1995	2010	1990	1995	2010	1990	1995	2010	1990	1995	2010
Average	1.76	1.76	1.63	0.74	0.73	0.61	1.3E-04	1.2E-04	1.1E-04	8.7E-05	5.9E-05	7.6E-05
Standard deviation	2.83	2.87	2.26	1.31	1.21	1.02	2.9E-04	2.7E-04	2.3E-04	1.7E-04	1.3E-04	1.4E-04

reduction plans as expressed by the individual European countries. The factors based on the 1995 emissions are chosen as the default EDIP2003 characterisation factors but the factors for the other years are shown to allow examination of the temporal sensitivity of the factors and, if wanted, to allow temporal differentiation for those emissions of the product system, which will take place in the future (e.g. from the late use stage of long-lived products or from the disposal stage). It is clear from Table 1 that for some countries, the site-dependent characterisation factors may vary considerably over time.

It is also observed from Table 1 that a few countries have negative characterisation factors for ozone exposure of human beings for either NO_x or NMVOCs in the 2010 scenario indicating that increased emissions will actually lead to reduced ozone formation. Applying a different model Krewitt and co-workers in their study of country specific damage factors for different air pollutants (Krewitt et al. 2001) have reported similar observations for particularly NO_x emissions from several European countries superimposed on a background of more moderate variations in the sensitivities among countries. The phenomenon demonstrates the often highly non-linear nature of the ozone formation. One could discuss whether it is preferable in life cycle impact assessment to apply a zero rather than negative characterisation factors in those few cases in order to refrain from rewarding countries for polluting.

The photochemical ozone formation is also influenced by meteorological conditions like solar radiation, wind speeds and directions and precipitation patterns. In order to reduce the influence of annual variations in the meteorological conditions, the characterisation factors for each of the emission years 1990, 1995 and 2010 were derived as the average of five different calculations using the meteorological data for the years 1989, 1990, 1992, 1993 and 1994 respectively.

2.3 Site-generic characterisation factors

The site-dependent characterisation factors of Table 1 are used to calculate the site-generic characterisation factors at the bottom of Table 1 as the emission-weighted average over all the countries and regions. The site-generic factors are compatible with the site-dependent factors in the sense that they are calculated using the same model and therefore cover the same part of the causality chain underlying photochemical ozone formation impacts. The only difference is that the spatial variation in the parameters of the model has been averaged out. The site-generic factors can be used in cases where spatial differentiation is unwanted or when the location of some of the processes in the product system is unknown or lies outside Europe. A standard deviation can be calculated for the site-generic factors reflecting the spatially determined variation inherent in them.

In contrast to the site-dependent factors, the site-generic factors only show minor temporal variations but the large standard deviations reflect the considerable variation in site-de-

pendent characterisation factors between countries for any emission year.

Emissions from a non-European or unknown region may, as a first approach, be characterised using the site-generic factors from Table 1. The standard deviations on the site-generic factors in these tables give a range of potential spatial variation for the application of the site-generic factor within Europe. Given the magnitude of the variation in emissions and sensitivities within Europe, the site-dependent factor is expected to lie within this range for most regions also in the rest of the world. Expert judgement may be used in the interpretation to assess whether the factor for emissions from processes in non-European regions should be found in the lower or upper end of the range.

The RAINS model only considers non-methane VOCs and NO_x. In comparison, the POCP values used in EDIP97 and other life cycle impact assessment methodologies are also available for CH₄ and carbon monoxide. To include carbon monoxide it was considered that, like many NMVOCs, it is a substance emitted from processes with incomplete combustion. It is also an intermediary product in the photochemical oxidation of some NMVOCs, and its atmospheric lifetime lies within the interval of non-methane VOCs. It was therefore assumed that the dispersion and deposition pattern of carbon monoxide is well represented by the model applied for NMVOCs, and in the calculation of photochemical ozone impact, carbon monoxide was hence treated as an NMVOC.

In contrast, methane is an extremely long-lived organic compound with an estimated residence time in the atmosphere of around 10 years. This means that a globally uniform distribution of methane in the atmosphere must be expected, and the contribution of methane to ozone formation is rather low at a regional level. In the calculation of characterisation factors it was thus decided to base the characterisation factors for methane on the site-generic factors developed for NMVOCs and correct for the fact that due to the long lifetime of methane, a large part of the ozone formed will expose ocean areas and hence not contribute to exposure of vegetation or humans. A correction factor of 0.5 was chosen, and the characterisation factor for methane is thus half the value of the site-generic characterisation factors for NMVOCs in Table 1. The uniform global distribution of methane means that the spatially determined standard deviation on the characterisation factor for methane is zero.

2.4 Use of the characterisation factors

Site-dependent characterisation factors can be applied right from the start in LCIA, but they can also be used to reduce uncertainty from a site-generic approach to acceptable levels. EDIP2003 adopts the latter approach. This means that first a site-generic characterisation is performed, and next the impacts from the main contributing processes are replaced by site-dependent characterisation up to a level where the calculated impact becomes stable (i.e. it can no longer change the conclusions of the LCA).

The site-generic photochemical ozone formation impacts of a product can be calculated according to the formula in Eq. 3:

$$\begin{aligned} \text{sgEP(po,hum)} &= \text{sg CF(po,hum)}_{\text{NO}_x} \cdot E_{\text{NO}_x} \\ &+ \text{sg CF(po,hum)}_{\text{VOC}} \cdot \sum_s \eta_s \cdot E_s + \text{sg CF(po,hum)}_{\text{CH}_4} \cdot E_{\text{CH}_4} \end{aligned} \quad (3)$$

Where:

sg EP(po,hum) is the site-generic photochemical ozone formation impact on human health expressed as persons exposed above threshold (in pers·ppm-hours/f.u.)

$\text{sg CF(po,hum)}_{\text{NO}_x}$ is the site-generic characterisation factor from Table 1 that relates emission of NO_x to the impacts on human health in the affected area (in pers·ppm-hours/g). The factor for 1995 is default.

$\text{sg CF(po,hum)}_{\text{VOC}}$ is the site-generic characterisation factor from Table 1 that relates emission of NMVOCs or CO to the impacts on human health in the affected area (in pers·ppm-hours/g). The factor for 1995 is default.

$\text{sg CF(po,hum)}_{\text{CH}_4}$ is the site-generic characterisation factor that relates emission of CH_4 to the impacts on human health in the affected area (in pers·ppm-hours/g). Taken as half the value for the NMVOCs in Table 1.

η_s is a substance-specific efficiency factor expressing the ozone creation potential of the individual volatile organic compound or CO relative to the ozone creation potential of the European average NMVOC (dimensionless). Efficiency factors for some of the more common NMVOCs and source-specified NMVOC mixtures are shown in Appendix 1 of the supporting material.

E_s is the emission of NO_x , CH_4 or individual or source-specified NMVOC or CO according to index (in g/f.u.)

A similar expression can be set up for calculation of the vegetation exposure to photochemical ozone.

The spatially determined variation which potentially lies hidden in the site-generic photochemical ozone impacts, can be estimated from the standard deviation given in Table 1 for each substance.

The photochemical ozone formation impact from a product is often dominated by a few processes. To avoid unnecessary work, applications where a site-dependent assessment is desired, may therefore start with calculation of the site-generic photochemical ozone formation impacts of the product as described above. Based on the site-generic impact, the processes with the dominating contributions can then be identified and their site-generic impacts be adjusted with the

relevant site-dependent characterisation factors according to Eq. 4. This procedure can be seen as a sensitivity analysis-based reduction of those uncertainties in the site-generic impact which are posed by refraining from site-dependent characterisation.

$$\begin{aligned} \text{sdEP(po,hum)}_p &= \text{sd CF(po,hum)}_{\text{NO}_x,i} \cdot E_{p,\text{NO}_x} \\ &+ \text{sd CF(po,hum)}_{\text{VOC},i} \cdot \sum_s \eta_{s,i} \cdot E_{p,s} + \text{sd CF(po,hum)}_{\text{CH}_4,i} \cdot E_{p,\text{CH}_4} \end{aligned} \quad (4)$$

Where:

sd EP(po,hum)_p is the site-dependent photochemical ozone formation impact on human health expressed as persons exposed above threshold by the selected process (p) (in pers·ppm-hours/f.u.)

$\text{sd CF(po,hum)}_{\text{NO}_x,i}$ is the site-dependent characterisation factor from Table 1 that relates emission of NO_x from country or region (i), where the selected process (p) is located, to the impacts on human health in the affected area (in pers·ppm-hours/g).

$\text{sd CF(po,hum)}_{\text{VOC},i}$ is the site-dependent characterisation factor from Table 1 that relates emission of NMVOCs or CO from country or region (i), where the selected process (p) is located, to the impacts on human health in the affected area (in pers·ppm-hours/g).

$\text{sd CF(po,hum)}_{\text{CH}_4,i}$ is the site-dependent characterisation factor from Table 1 that relates emission of CH_4 to the impacts on human health in the affected area (in pers·ppm-hours/g). Taken as half the site-generic value for the NMVOCs.

$\eta_{s,i}$ is a substance-specific efficiency factor expressing the ozone creation potential of the individual volatile organic compound or CO (s) relative to the ozone creation potential of the European average NMVOC (dimensionless). See Appendix 1 of the supporting material.

$E_{p,s}$ is the emission of NO_x , CH_4 or individual or source-specified NMVOC or CO (s), according to index, from process (p) (in g/f.u.)

... and similarly for calculation of the site-dependent vegetation exposure to photochemical ozone.

Example. The EDIP2003 characterisation factors are demonstrated by a case comparing two structural components in the seat of an office chair. One is made from flow-injection moulded plastic (20 g PE), the other from die-cast zinc (50 g zinc) (for the full case description see Hauschild and Potting, 2005). First, the site-generic impacts are calculated according to equation (3) applying the site-generic characterisation factors from Table 1 and the substance-specific

Table 2: Site-generic photochemical ozone impacts on vegetation and human health for one supporting block made from plastic or zinc (mean and standard deviation representing spatial variation)

	Site-generic ozone impact, vegetation		Site-generic ozone impact, human health	
	$m^2 \text{ppm-hours/f.u.}$		$\text{pers} \cdot \text{ppm-hours/f.u.}$	
	mean	std.dev.	mean	std.dev.
Zinc component	13.4	21.7	9.0E-04	1.9E-03
Plastic component	7.4	12.3	5.2E-04	1.0E-03

efficiency factors for different NMVOCs and CO from Annex 1 of the supporting material. The results are shown in Table 2.

The zinc component has the largest impacts in both categories. However, the potential spatial variation is so large (as revealed by the spatially determined standard deviation) that the conclusion is highly uncertain. Therefore, a site-dependent characterisation is performed for those processes which contribute the most to the site-generic impacts, in order to reduce the spatially determined uncertainty and strengthen the conclusion.

The impacts on vegetation as well as on human health are dominated by NO_x contributions, and an emission of unspecified NMVOCs is also noticeable. The main NO_x sources for the zinc component, are identified as the production of zinc from ore which takes place in Bulgaria, the casting of the component which takes place in former Yugoslavia and that leg of the transport of the component, which takes place by truck through Germany. For the plastic component, the main sources for NO_x are found to be the production of plastic polymer in Italy, the flow injection moulding of the supporting block in Denmark and the transportation of the component by truck, again mainly through Germany. The unspecified NMVOC-emission from the plastic component is caused by the production of the plastic polymer in Italy, and for the zinc component, it comes from the casting process in Yugoslavia. The emissions from these processes together contribute more than 99% for the zinc component and 80% for the plastic component to the impacts on vegetation and on human health.

In the calculation of the site-dependent impacts for these key processes according to equation (4), the relevant site-dependent factors from Table 1, and the efficiency factors from Appendix 1 of the supporting material, are applied. The site-generic impacts from the key processes are subtracted from the original site-generic impacts in Table 2 by applying equation (3), and the site-dependent impacts from the key processes are added. The photochemical ozone impacts thus corrected are found in Table 3 together with the residual spatially determined uncertainty, and the differences to the site-generic impacts of Table 2 are illustrated in Fig. 1.

For photochemical ozone impact on vegetation, more than 99% of the impacts for the zinc component now include the spatial information, and the standard deviation representing the residual spatially determined uncertainty is less than 1% (data not shown). The spatially conditioned uncertainty of the impact has thus largely been cancelled, and even if the site-dependent characterisation was performed for all the remaining processes in the product system, the result would not change significantly. For the plastic component, about 90% of the impacts on vegetation and human health include the spatial information (data not shown). As seen from the residual spatial uncertainty for the plastic component in Table 3, there is still a considerable spatially determined uncertainty left, and this means that for the plastic component, the figures for impacts on vegetation and particularly on human health might still change if further spatial characterisation were performed. More key processes would thus need to be included in order to cancel the spatially determined uncertainty of the conclusion.

Table 3: Photochemical ozone impacts from the two product systems after site-dependent characterisation of key process emissions

	Ozone impact, vegetation	Ozone impact, human health
	$m^2 \text{ppm-hours/f.u.}$	$\text{pers} \cdot \text{ppm-hours/f.u.}$
Zinc component	17.5	8.0E-04
- residual spatial uncertainty	0.12	0.12E-04
Plastic component	9.1	6.5E-04
- residual spatial uncertainty	2.8	2.6E-04

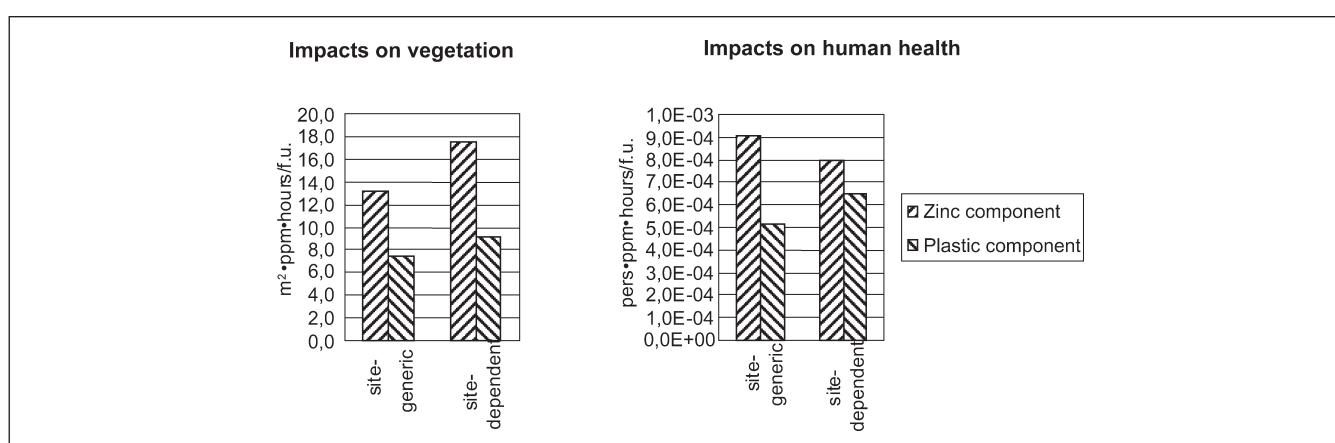


Fig. 1: Site-generic and site-dependent photochemical ozone impacts on vegetation and human health from the product systems of the two components

As seen from Fig. 1, the inclusion of spatial differentiation, at the level of emission country, strengthens the dominance of the zinc component in ozone impact on vegetation. For ozone impacts on human health, on the other hand, the two alternatives become very similar when a major part of the spatial variation in the dispersion patterns and sensitivity of the exposed environment (i.e. population density) is eliminated. Even though only 10% of the site-generic impact score has not been included in the spatial characterisation, the residual spatially determined uncertainty is, however, still considerable for the plastic component, as seen from Table 3. Inclusion of further emissions (mainly NO_x) may thus still alter the results for this component.

3 Discussion and Conclusions

The example illustrates the inclusion of spatial variation in the characterisation of photochemical ozone impacts. The EDIP2003 factors make this possible for processes located in Europe. Together with other related work on spatial differentiation in the characterisation modelling for photochemical oxidants (Bare et al. 2003, Krewitt et al. 2001), the EDIP2003 factors hold the following advantages over the POCP factors which are traditionally applied in LCIA:

- The contribution of NO_x to photochemical ozone formation can be included in the impact potentials. Normalisation references were calculated based on the total predicted impacts from all European emissions in the reference year 1995 (data not shown). The normalisation references show that the contribution from NO_x is around 2/3 of the total ozone formation impact in Europe (Hauschild et al. 2005).
- From Table 1 it is seen that in most cases the characterisation factor for NO_x is higher than the characterisation factor for NMVOCs. The significance of this novelty depends for a specific product system on the quantities of NO_x and NMVOCs emitted.
- The site-dependent characterisation factors in Table 1 for exposure of humans to photochemical ozone show a variation between countries of up to three orders of magnitude. In comparison, the variation among individual substances of the POCP factors which traditionally are applied in the characterisation of photochemical ozone formation, is within one order of magnitude. It thus seems more important to represent the spatially determined variation in human exposure than the difference in photochemical ozone creation potential among the substances.
- Nonetheless efficiency factors which represent the substance-specific variation in the ozone creation potential of individual VOCs are provided in Annex 1 of the supporting material. The factors have been derived from the POCP values for areas with a high background concentration of NO_x , but if there is a wish to represent the substance-specific variation in ozone formation potential, it is recommended to apply them also for emissions taking place in low NO_x regions as the variation between high- and low NO_x POCP factors for one substance is modest compared to the variation between the POCPs of different substances (Hauschild and Wenzel 1998).

- The impact potential calculated with the EDIP2003 factors is more straightforward to interpret in terms of environmental damage since it is modelled further along the impact chain to include exposure of human beings and vegetation instead of just predicting the potential formation of ozone. Although still a midpoint method, EDIP2003 moves the category indicator far in the direction of endpoint.
- The EDIP2003 factors are calculated for the emission levels of three different years (1990, 1995 and predicted emission levels for 2010) for average meteorological conditions which allows judging their sensitivity to this temporal variation.

The site-dependent characterisation also introduces some new challenges:

- In traditional midpoint approaches, all environmental impacts from photochemical ozone formation have been represented by one impact category defined early in the impact pathway. Now we must operate with two (sub)categories. Since large parts of their environmental mechanisms are identical, there may be a wish to aggregate them into one category. This will require development of weighting factors reflecting their relative importance. It may be stated, that this weighting was inherent in the POCP-based approach, and now it just has to be made explicit.
- Tropospheric ozone impacts on man-made materials are not explicitly addressed by the site-dependent factors. In the POCP-based approach they were implicitly included together with all other impacts of the tropospheric ozone. In the EDIP2003 approach they may also be seen as covered together with human health impacts since their distribution and hence exposure follows the distribution of humans. This assumption must influence the weight this (sub)category is given in an eventual weighting against other environmental impact categories.
- The life cycle of a product is normally global, and while the site-generic EDIP2003 factors may be applied for emissions occurring outside Europe, site-dependent characterisation factors for other continents are needed. Indeed, important LCIA method development activities in USA and Japan are very aware of the relevance of spatial differentiation (Bare et al. 2003, Itsubo and Inaba 2003). A comparison and possibly an intercalibration of the different approaches to site-dependent characterisation of photochemical ozone as well as other non-global impacts is a pressing future research topic.
- In order to make application of site-dependent characterisation practically available to LCA practitioners, the site-dependent characterisation factors must be included in PC tools used for life cycle assessment. A pre-requisite is that the geographical location of processes, which is often used anyway to select process technologies and to quantify transport distances, is recorded in the life cycle databases of the tools. Furthermore, the inventory must be available for characterisation in a non-aggregated form. The traditional reporting of an aggregated inventory loses the geographical information of the individual emissions, which is needed for site-dependent characterisation.

When to use spatial differentiation in characterisation modelling? The relevance of spatial differentiation depends on the goal of the study. For many applications of LCA, there is a general requirement that the impact assessment should give a best estimate of the environmental impacts that are caused by the emissions from the product system. If the spatial variation is important at the scale of countries, and if the uncertainty of the applied models is acceptable compared to the reduction they can give in the spatially determined uncertainty, spatial differentiation will thus improve the characterisation modelling.

There are also applications of LCA, where the information provided through inclusion of spatial differentiation may not be relevant to the goal of the study. This can be the case for preparation of environmental product declarations and ecolabel criteria where there may be a politically founded wish to avoid different treatment of different regions on grounds of their environmental robustness.

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Appendix: Supporting information

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Supporting Information**Annex 1:** Efficiency factors for individual VOCs and source-specified VOCs

The ozone formation model applied in RAINS does not allow distinction between individual VOCs. This eliminates a source of variation in the factors but since the variation between the ozone formation potentials lies within a factor 2–4 for most VOCs, this potential error is small compared to the error eliminated by including the contribution from NO_x. Nonetheless, it is suggested to introduce differentiation between the individual VOCs through an efficiency factor η_s representing the ozone formation potential of a specific VOC (s) relative to the average ozone formation potential of European VOC emissions

Table S.I.4: The dimensionless efficiency factor is calculated as the quotient between the respective POCP-factors for 4–9 days in high NO_x-areas (the EDIP97 characterisation factors for high NO_x-areas, Wenzel et al. 1997) and the European average POCP

Individual VOCs and methane	Efficiency factor η_s	Individual VOCs, carbon monoxide and source-specified VOCs	Efficiency factor η_s
Alkanes	1.0	Aldehydes	1.5
ethane	0.21	formaldehyde	1.1
propane	1.1	acetaldehyde	1.3
n-butane	1.0	propionaldehyde	1.5
isobutane	0.79	butyraldehyde	1.4
n-pentane	1.0	isobutylaldehyde	1.6
isopentane	0.74	valeraldehyde	1.7
n-hexane	1.1	acrolein	2.0
2-methylpentane	1.3	Ketones	1.0
3-methylpentane	1.1	acetone	0.45
2,2-dimethylbutane	0.63	methyl ethyl ketone	1.1
2,3-dimethylbutane	0.96	methyl isobutyl ketone	1.6
n-heptane	1.3	Alcohols	0.66
2-methylhexane	1.2	methanol	0.31
3-methylhexane	1.2	ethanol	0.67
n-octane	1.2	isopropanol	0.50
2-methylheptane	1.2	butanol	1.0
n-nonane	1.2	isobutanol	0.75
2-methyloctane	1.3	butan-2-diol	0.75
n-decane	1.2	Ethers	1.0
2-methylnonane	1.1	dimethyl ether	0.75
n-undecane	1.1	propylene glycol methyl ether	1.3
n-dodecane	1.0	Esters	0.51
Alkenes	2.2	methyl acetate (=dimethyl ester)	0.06
ethylene	2.5	ethyl acetate	0.55
propylene	2.6	isopropyl acetate	0.54
1-butene	2.4	n-butyl acetate	0.81
2-butene (trans)	2.5	isobutyl acetate	0.83
Isobutene	1.5	propylene glycol methyl ether acetate	0.25
2-pentene (trans)	2.3	Chloro-alkanes	0.011
1-pentene	2.6	methylene chloride	0.023
2-methylbut-1-ene	1.9	chloroform	0.0075
3-methylbut-1-ene	2.2	methyl chloroform	0.0025
2-methylbut-2-ene	1.9	Chloro-alkenes	0.64
2-methylpropene	1.6	trichloroethylene	0.17
Isoprene	2.0	tetrachloroethylene	0.01
Alkynes		allyl chloride	1.8
acetylene	0.42	Inorganic compounds	
Aromatics	1.9	carbon monoxide	0.075
benzene	0.47	Source-specified VOC mixtures	
toluene	1.4	Petrol-powered car, exhaust	1.5
o-xylene	1.7	Petrol-powered car, vapour	1.3
m-xylene	2.5	Diesel-powered car, exhaust	1.5
p-xylene	2.2	Power plants	1.3
ethylbenzene	1.5	Combustion of wood	1.5
n-propylbenzene	1.2	Food industry	1.0
isopropylbenzene	1.4	Surface coating	1.3
1,2,3-trimethylbenzene	2.9	Chemical cleaning of clothes	0.75
1,2,4-trimethylbenzene	3.0	Refining and distribution of oil	1.3
1,3,5-trimethylbenzene	2.9	Natural gas leakage	0.050
o-ethyltoluene	1.7	Coal mining	0.018
m-ethyltoluene	2.0	Farming	1.0
p-ethyltoluene	1.8	Landfilling of household waste	0.018

$$\eta_s = \frac{POCP_s}{POCP_{\text{European average}}}$$

The European average POCP was determined for 1985 emissions at 0.40 in Hauschild and Wenzel (1998). Based on this figure and the POCP values applied for characterisation factors in EDIP97, the efficiency factor is calculated and tabulated below for the most common individual VOCs as well as for source-specified VOC-mixtures frequently encountered in life cycle inventories.